

1 Neighborhood-level nitrogen dioxide inequalities
2 contribute to surface ozone variability in Houston,
3 Texas

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23 **Abstract.** In Houston, Texas, nitrogen dioxide (NO_2) air pollution disproportionately affects
24 Black, Latinx, and Asian communities, and high ozone (O_3) days are frequent. There is limited
25 knowledge of how NO_2 inequalities vary in urban air quality contexts, in part from the lack of
26 time-varying neighborhood-level NO_2 measurements. First, we demonstrate that daily TROPOMI
27 NO_2 tropospheric vertical column densities (TVCDs) resolve a major portion of census tract-scale
28 NO_2 inequalities in Houston, comparing NO_2 inequalities based on TROPOMI TVCDs and
29 spatiotemporally coincident airborne remote sensing (250 m \times 560 m) from the NASA TRacking
30 Aerosol Convection ExpeRiment–Air Quality (TRACER-AQ). We further evaluate the
31 application of daily TROPOMI TVCDs to census tract-scale NO_2 inequalities (May 2018–
32 November 2022). This includes explaining differences between mean daily NO_2 inequalities and
33 those based on TVCDs oversampled to $0.01^\circ \times 0.01^\circ$ and showing daily NO_2 column-surface
34 relationships weaken as a function of observation separation distance. Second, census tract-scale
35 NO_2 inequalities, city-wide high O_3 , and mesoscale airflows are found to covary using principal
36 component and cluster analysis. A generalized additive model of O_3 mixing ratios versus NO_2
37 inequalities reproduces established nonlinear relationships between O_3 production and NO_2
38 concentrations, providing observational evidence that neighborhood-level NO_2 inequalities and O_3
39 are coupled. Consequently, emissions controls specifically in Black, Latinx, and Asian
40 communities will have co-benefits, reducing both NO_2 disparities and high O_3 days city wide.

41 **Synopsis.** Most neighborhood-level NO₂ inequalities can be observed with daily TROPOMI
42 observations; the unequal NO₂ distribution affects O₃ chemistry in Houston, Texas.

43 **Keywords.** Nitrogen dioxide, ozone, TROPOMI, urban air pollution, environmental racism

44 1 **INTRODUCTION**

45 Houston, Texas is a large U.S. city and center for petrochemical refining that faces multiple air
46 quality challenges. Historical and contemporary policies and practices continue to
47 disproportionately offload the environmental costs of industry and transportation on Black, Latinx,
48 and Asian communities,^{1, 2} causing measurable inequalities in the distribution of nitrogen dioxide
49 (NO₂) and other primary pollutants.³⁻⁹ Houston is also currently ranked among the top-ten most
50 ozone (O₃) polluted cities in the U.S., with residents experiencing frequent exceedances of health-
51 based O₃ standards city wide.¹⁰ Recent analytical advances have produced more spatially detailed
52 descriptions of neighborhood-level urban air pollution inequalities,¹¹⁻¹⁵ including for NO₂.¹⁶⁻¹⁸
53 However, enhanced spatial information has generally relied on time-averaged and/or short-
54 duration observations, representing conditions that potentially infrequently occur and limiting our
55 understanding of relationships between NO₂ inequalities and broader urban air quality issues such
56 as O₃. This has policy relevance as states have regulatory authority around O₃ compliance that they
57 often lack or decline to use regarding air pollution environmental injustice.

58 NO₂ is a criteria pollutant regulated by the U.S. Environmental Protection Agency (EPA). NO₂ is
59 a primary pollutant (or pseudo-primary pollutant) with a summertime atmospheric lifetime as short
60 as a few hours. Primary pollutants are highly spatiotemporally variable, exhibiting atmospheric
61 dispersion gradients of hundreds of meters to 1–2 km.^{11, 19, 20} NO₂ is emitted as NO_x (\equiv NO + NO₂),
62 with vehicles and electricity generation being major NO_x sources in U.S. cities.²¹⁻²³ Houston is

63 also a global hub for petrochemical manufacturing, where refineries and industrial activities
64 contribute a large portion of NO_x emissions,²⁴⁻²⁶ especially in the Houston Ship Channel,²⁴⁻²⁶ a
65 residential and industrial area along the Buffalo Bayou River, connecting downtown to Galveston
66 Bay and the Gulf of Mexico (Figure 1). Associated with numerous adverse²⁷⁻³¹ and unequal health
67 impacts,²⁸ NO₂ is a common proxy for toxic combustion and traffic air pollution mixtures in health
68 studies.³² High-volume roadways and heavy-duty diesel truck traffic overburden communities of
69 color,^{33, 34} and living near roadways is linked to asthma-related urgent medical visits, pediatric
70 asthma, preeclampsia and preterm birth, and cardiac and pulmonary mortality.³⁵⁻⁴⁰

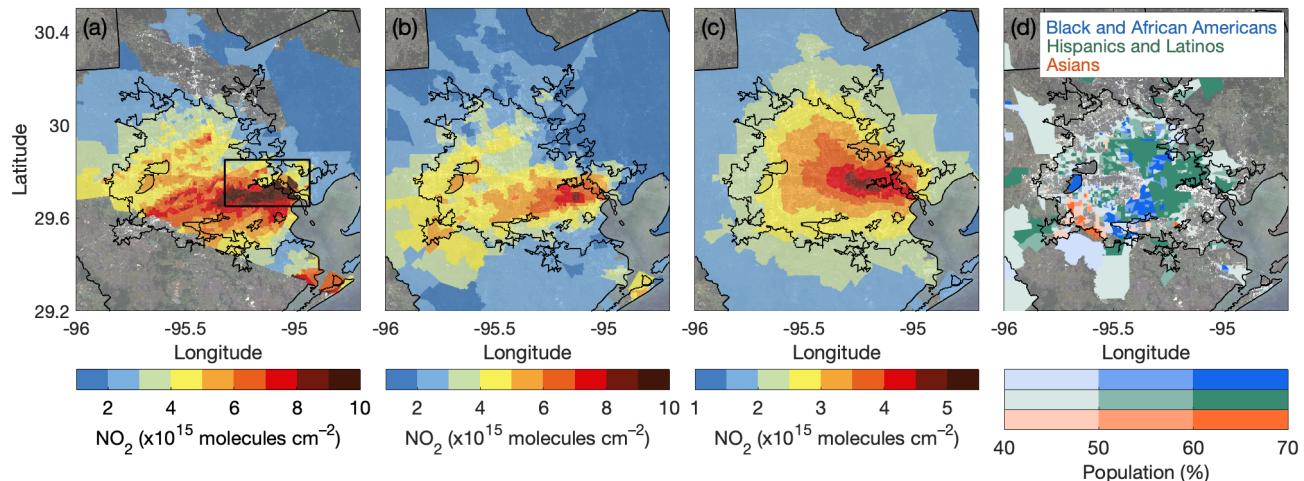
71 Neighborhood-level NO₂ inequalities with race and ethnicity can be observed from space using
72 the TROPOspheric Monitoring Instrument (TROPOMI).^{3, 16, 41-45} This was first demonstrated by
73 Demetillo et al.,³ who showed relative census tract-scale NO₂ inequalities based on TROPOMI
74 tropospheric vertical column densities (TVCDs) oversampled to $0.01^\circ \times 0.01^\circ$ agreed with results
75 from fine-scale (250 m \times 500 m) airborne remote sensing during the NASA Deriving Information
76 on Surface Conditions from COlumn and VERtically Resolved Observations Relevant to Air
77 Quality (DISCOVER-AQ) in Houston. In addition, spatial patterns in oversampled TROPOMI
78 TVCDs reflected NO₂ distributions at the surface, a conclusion based on comparisons with in-situ
79 aircraft NO₂ vertical profiles from DISCOVER-AQ and surface measurements.³ In a subsequent
80 analysis of 52 U.S. cities, Demetillo et al.¹⁶ reported oversampled TROPOMI NO₂ inequalities
81 were invariant with urban racial segregation structure,³⁴ meaning that TROPOMI resolves inter-
82 tract NO₂ differences even when segregated tracts do not spatially aggregate into larger regions.
83 Dressel et al.⁴¹ found mean daily TROPOMI observations (3.5 km \times 5.5 km at nadir) without
84 oversampling also captured a majority of tract-scale NO₂ inequalities compared to fine-scale (250
85 m \times 250 m) airborne remote sensing and agreed with relative NO₂ inequalities based on TVCDs

86 oversampled to $0.01^\circ \times 0.01^\circ$ to within associated uncertainties, at least in New York City, New
87 York and Newark, New Jersey. Daily NO_2 inequalities, when uncertainties are well-characterized,
88 can be analyzed statistically and situated within our broader understanding of urban air quality.⁴¹

89 NO_2 is an O_3 precursor and temporary O_3 reservoir ($\text{O}_x \equiv \text{NO}_2 + \text{O}_3$), with O_3 production chemistry
90 varying nonlinearly with NO_2 and the reactivity of volatile organic compounds (VOCs) with
91 hydroxyl radical (OH). O_3 pollution in Houston is attributed in large part to the combination of
92 high NO_x and reactive VOC emissions by industries in the Ship Channel and gulf breeze
93 airflows.^{26, 46-51} While O_3 air quality has improved,⁵²⁻⁵⁴ exceedances of the health-based maximum
94 daily average 8-h (MDA8) O_3 National Ambient Air Quality Standard (NAAQS) of 70 ppb are
95 frequent, with 141 exceedance days in the Houston Metropolitan Statistical Area (MSA) over May
96 2018–November 2022 (our study period). O_3 is a secondary and intermediately long-lived
97 pollutant. As a result, O_3 exhibits less intraurban heterogeneity than NO_2 and is not generally
98 associated with neighborhood-level disparities.⁵⁵ However, because NO_2 and VOC concentrations
99 are spatiotemporally variable, O_3 production (PO_3) chemistry is as well,⁵⁶⁻⁵⁸ with NO_2 inequalities
100 and city-wide O_3 potentially coupled. In Houston, the largest NO_2 inequalities during DISCOVER-
101 AQ corresponded to a severe O_3 event with MDA8 O_3 of 124 ppb (LaPorte Sylvan Beach, 25
102 September 2013).³ In New York City–Newark, tract-scale NO_2 inequalities were positively
103 associated with summertime MDA8 O_3 (2018–2021), with Spearman correlation coefficients of
104 0.41–0.55 for different population groups.⁴¹

105 Here, we describe census tract-scale TROPOMI NO_2 inequalities and investigate relationships
106 with MDA8 O_3 in Houston. As a first step, we evaluate daily TROPOMI NO_2 inequalities with
107 race-ethnicity, advancing our understanding of the application of mean daily TROPOMI NO_2
108 TVCDs to NO_2 inequalities developed in New York City–Newark.⁴¹ We compare daily TROPOMI

109 NO₂ inequalities against measurements of spatiotemporally coincident airborne remote sensing
 110 (250 m × 560 m) during the NASA TRacking Aerosol Convection ExPeRiment–Air Quality
 111 (TRACER-AQ) in September 2021, discuss differences between relative and absolute mean daily
 112 and oversampled TROPOMI NO₂ inequalities, and present column-surface relationships as a
 113 function of measurement separation distance and surface wind conditions. Second, we statistically
 114 analyze TROPOMI NO₂ inequalities (May 2018–November 2022), interpreting covariations
 115 between neighborhood-level NO₂ inequalities, overall NO₂ pollution, and urban O₃ air quality in
 116 ways that have policy implications.



117
 118 **Figure 1.** Example of census tract-scale GCAS NO₂ columns (molecules cm⁻²) collected on 25
 119 September 2021 at 2–5 pm (a), TROPOMI TVCDs on the same day, with a mean pixel size of 21
 120 ± 0.6 km² (b), and oversampled TROPOMI TVCDs (0.01° × 0.01°) over May 2018–November
 121 2022 (c). Also shown, the percent population for the largest race-ethnicity group in each census
 122 tract for Black and African Americans (blue), Hispanics and Latinos (green), and Asians (orange)
 123 (d). The inner and outer black lines are the Urbanized Area (UA) and Metropolitan Statistical Area
 124 (MSA) boundaries, respectively. The thick black box is the Houston Ship Channel (a). Background
 125 map data: Landsat 8 composite (January 2017–June 2018). Corresponding wind conditions are
 126 presented in Figure S1.

127 2 MEASUREMENTS AND METHODS

128 **TROPOMI.** TROPOMI is a hyperspectral spectrometer onboard the sun-synchronous European
 129 Space Agency Copernicus Sentinel-5 Precursor (S-5P) satellite.^{59,60} NO₂ is retrieved by fitting the

130 405–465 nm spectral band based on an updated Dutch OMI (Ozone Monitoring Instrument) NO₂
131 (DOMINO) algorithm and work from the Quality Assurance for Essential Climate Variables
132 project.^{61–65} NO₂ observations are converted to TVCDs via an air mass factor (AMF), which relies
133 on spatially and temporally coarse inputs, e.g., clouds, surface albedo, and NO₂ profile shape, that
134 can bias NO₂ TVCDs low under high NO₂ conditions.⁶⁶ The application of TROPOMI NO₂
135 TVCDs to census tract-scale NO₂ inequalities has been evaluated through comparison with
136 airborne remote sensing that resolves NO₂ distance decay gradients, both in terms of TVCDs first
137 oversampled to $0.01^\circ \times 0.01^\circ$ ³ and daily TVCDs,⁴¹ with TROPOMI capturing similar relative but
138 lower absolute population-weighted census tract-scale NO₂ inequalities. While the sensitivity of
139 TROPOMI is lower near the surface,^{67, 68} there are no physical processes in the free troposphere
140 that maintain intraurban gradients corresponding to neighborhood-level race-ethnicity. TROPOMI
141 TVCDs have been shown to reflect intraurban spatiotemporal NO₂ variability at the surface, a
142 critical analytical requirement for informing decision making around environmental racism.^{3, 16, 41}
143 Based on 144 in-situ NO₂ vertical profiles throughout Houston from DISCOVER-AQ, Demetillo
144 et al.³ reported that the slope of the linear fit between the measured full column (extending up to 3
145 km) and NO₂ column within the convective boundary layer was 0.98 ± 0.15 ($r = 0.99$), with no
146 significant location-specific differences. Multiple authors have shown TROPOMI and OMI NO₂
147 TVCDs correlate with surface-level nitrogen dioxide (NO₂^{*}) measurements and, more
148 importantly, that correlation coefficients decrease with increasing spatial separation between
149 columns and monitors on the scales of NO₂ spatial variability.^{3, 16, 41, 69}

150 From 1 May 2018 to 5 August 2019, the TROPOMI nadir spatial resolution was $3.5 \text{ km} \times 7 \text{ km}$;
151 from 6 August 2019 to present, the nadir spatial resolution improved to $3.5 \text{ km} \times 5.5 \text{ km}$.⁷⁰ The S-
152 5P satellite crosses the equator at ~1:30 pm local time (LT) and overflies Houston at 12–3 pm LT,

153 typically once but occasionally twice daily. When there are two TROPOMI overpasses over
154 Houston on the same day, we use the first overflight only. We use current Level 2 NO₂ TVCDs
155 (version 02.04.00) with quality assurance values >0.75, as recommended,⁷¹ from operationally
156 reprocessed (RPRO, collection identified: ‘03’, 1 May 2018–25 July 2022) and offline (OFFL, 26
157 July 2022–30 November 2022) products. A key update in version 02.04.00 is the use of a surface
158 albedo climatology derived from TROPOMI observations rather than the coarse spatial resolution
159 OMI surface albedo climatology (0.5° × 0.5°).⁷¹ TROPOMI NO₂ inequalities can be sensitive to
160 product version; for example, Dressel et al.⁴¹ found census tract-scale NO₂ inequalities based on
161 NO₂ TVCDs reprocessed on the S-5P Products Algorithm Laboratory (S5P-PAL) system were 3–
162 6 points (10–20%) higher over the New York City–Newark urbanized area (UA) than those
163 computed using a then current version of operational product (version 01.02.02). We compared
164 NO₂ inequalities using version 02.04.00 (RPRO) and S5P-PAL reprocessed TVCDs over January–
165 December 2019 but find results were statistically indistinguishable.

166 **GCAS.** The Geostationary Coastal and Air Pollution Events (GEO-CAPE) Airborne Simulator
167 (GCAS) makes hyperspectral nadir-looking measurements of backscattered solar radiation in the
168 ultraviolet and visible in two channels at wavelengths 300–490 nm (optimized for air quality) and
169 480–900 nm (optimized for ocean color).⁷² Each channel uses a two-dimensional (2D) charge-
170 coupled device (CCD) array detector, where one CCD dimension provides spectral coverage and
171 the other the cross-track spatial coverage across a ~45° field of view in the air quality channel.
172 GCAS was developed as a technology-demonstration instrument for the GEostationary Coastal
173 and Air Pollution Events (GEO-CAPE) decadal survey and functions as a satellite analog in NASA
174 airborne research. GCAS NO₂ column retrievals are validated over urban areas and consist of a
175 two-step approach similar to algorithms used for other major satellite instruments, including

176 TROPOMI.⁷³⁻⁷⁵ Briefly, NO₂ differential slant columns are retrieved fitting across 425–460 nm
177 using the QDOAS spectral fitting package⁷⁶ and a reference spectrum measured at a nearby
178 location away from NO_x emissions sources. The AMF is largely a function of viewing and solar
179 geometries, surface reflectance, and atmospheric and trace gas vertical profiles.^{73, 77} GCAS
180 retrievals for TRACER-AQ use the NASA GEOS-CF model analyses (0.25° × 0.25°).⁷⁸ Other
181 components of the retrieval follow Judd et al.,⁷⁷ where column uncertainties over New York City–
182 Newark were ±25% and unbiased compared to coincident Pandora measurements, ground-based
183 total NO₂ columns with relatively low uncertainties from AMFs that do not vary with NO₂ vertical
184 profile shape or surface albedo.⁷⁹ During TRACER-AQ, GCAS NO₂ columns were averaged to
185 250 m (cross-track) × 560 m (along track). GCAS flew onboard the NASA Johnson Space Center
186 Gulfstream V (JSC GV) research aircraft on 11 days in September 2021. We use measurements
187 from the 27 cloud-free flights sampling at least 60% of census tracts in the Houston MSA (Table
188 S1). GCAS flew a repeated flight pattern in the morning (~9–11:30 am LT), midday (~11:30 am–
189 2 pm LT), and afternoon (~2:30–5 pm LT), sampling 83 ± 4% ($\pm 1\sigma$) of tracts with similar, but not
190 identical, demographics to the MSA (Tables S2–S3).

191 **Surface NO₂*, O₃, and Meteorological Measurements.** NO₂* observations are collected at 23
192 stations across the MSA (Figure S2a) and provided through the U.S. EPA Air Quality System.⁸⁰
193 NO₂* is mostly measured by decomposing NO₂ to NO over a heated molybdenum catalyst and
194 detecting NO by chemiluminescence, a technique with a known positive interference from other
195 nitrogen compounds, which also thermally decompose across the catalyst at non-unity
196 efficiency.⁸¹⁻⁸³ The term NO₂* acknowledges this interference, which, while affecting accuracy,
197 has a smaller effect on precision.⁸⁴ Two stations in the MSA are near-roadway monitors. We use
198 O₃ mixing ratios measured at 21 stations, many of which also house NO₂* instruments (Figure

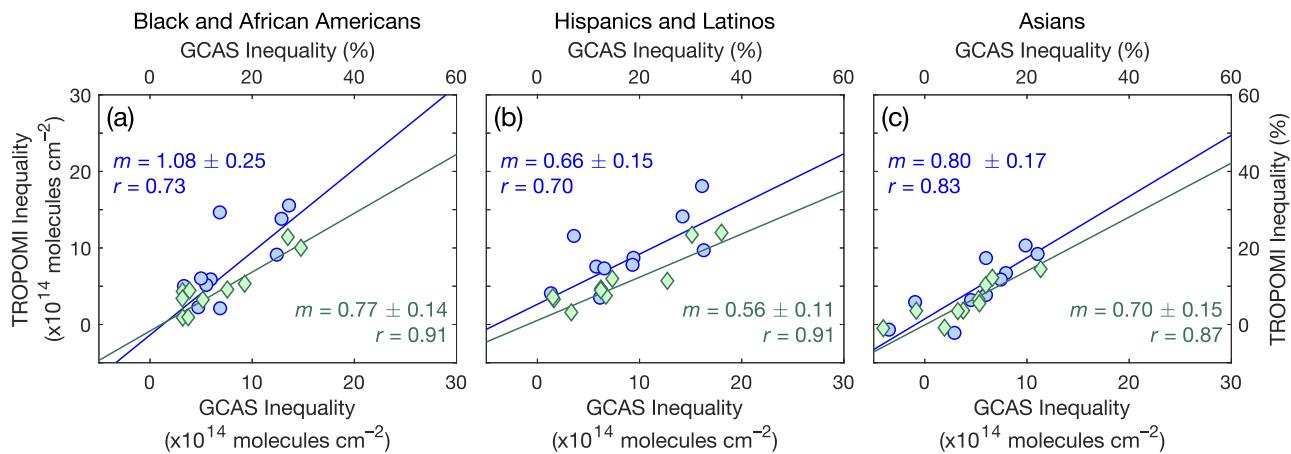
199 S2b), converted to MDA8 O₃. We use 1-h measurements of wind speed (resultant), wind direction,
200 and air temperature and daily maximum temperatures collected at 23 stations (Figure S2c) with
201 observations on at least 50% of days during O₃ season, defined in Houston as March–November,⁸⁵
202 when MDA8 O₃ NAAQS exceedances are most likely to occur.

203 **Census Tract-Scale Inequalities.** We calculate area-weighted mean NO₂ TVCDs within 2020
204 census tract polygons across the Houston UA and MSA and population weight tract-average
205 TVCDs using race and ethnicity data from the U.S. Census 5-year 2020 American Community
206 Survey (ACS). The ACS subsamples census unit populations and applies a complex weighting
207 process to account for variability in tract-level sampling rates and differential group response rates.
208 The weighting process prioritizes accuracy over precision, which we manage using population-
209 weighting and aggregation across the UA and MSA.^{86, 87} Tract-scale NO₂ inequalities with race-
210 ethnicity are reported as relative (%) and absolute (molecules cm⁻²) differences between
211 population-weighted NO₂ TVCDs (eq. S1^{3, 18, 88}) for non-Hispanic/Latino Black and African
212 Americans, Hispanics and Latinos of all races, and non-Hispanic/Latino Asians compared to non-
213 Hispanic/Latino whites in tracts with populations equal to or greater than the mean across tracts
214 with observations. NO₂ differences with race and ethnicity are treated as a proxy for racism.

215 **3 RESULTS AND DISCUSSION**

216 **Evaluating Daily TROPOMI NO₂ Inequalities in Houston, Texas.** We first compare spatially
217 and temporally coincident daily census tract-scale TROPOMI NO₂ inequalities against those
218 computed using GCAS NO₂ columns, which have sufficient spatial resolution to observe NO₂
219 dispersion gradients. Correspondence between daily TROPOMI and GCAS inequalities is
220 described using Pearson correlation coefficients and slopes derived from an unweighted bivariate

221 linear regression of simultaneous observations, defined as occurring within ± 30 minutes (Figure
 222 2). TROPOMI and GCAS NO₂ inequalities are strongly correlated, with r values of 0.70–0.83
 223 (relative) and 0.87–0.91 (absolute), indicating daily TROPOMI NO₂ TVCDs reflect the variability
 224 of spatially detailed GCAS observations day to day. Regression slopes are 0.66 ± 0.15 to $1.08 \pm$
 225 0.25 for relative and 0.56 ± 0.11 to 0.77 ± 0.14 for absolute inequalities; therefore, daily TROPOMI
 226 NO₂ TVCDs capture a major portion of tract-scale inequalities in Houston. Slopes for relative
 227 inequalities are larger than for absolute inequalities, with relative differences easier to distinguish
 228 using measurements coarser than distance decay gradients. This is consistent with results from
 229 daily observations in New York City–Newark⁴¹ and reinforces conclusions based on oversampled
 230 TVCDs in Houston by Demetillo et al.,³ where TROPOMI resolved comparable relative but lower
 231 absolute inequalities than GCAS during DISCOVER-AQ.



232
 233 **Figure 2.** Spatiotemporally coincident (± 30 min) relative (%) (blue circles) and absolute
 234 (molecules cm^{-2}) (green diamonds) GCAS and TROPOMI NO₂ inequalities during TRACER-AQ
 235 for Black and African Americans (a), Hispanics and Latinos (b), and Asians (c) in comparison to
 236 non-Hispanic/Latino whites with slopes (m), based on an unweighted bivariate linear regression,
 237 and Pearson correlation coefficients (r) of relative (blue) and absolute (green) inequalities.
 238 We test the sensitivity of daily TROPOMI census tract-scale NO₂ inequalities to TROPOMI
 239 observation spatial resolution by comparing NO₂ inequalities across the natural variability in daily

240 mean TROPOMI pixel size, ranging 20–89 km² with a mean of 39 ± 16 km² ($\pm 1\sigma$ standard
241 deviation) UA wide (May 2018–November 2022). Because daily inequalities are sensitive to
242 observation coverage, we first remove days with NO₂ observations in fewer than 20% of tracts in
243 the domain (discussed below). We group observations according to thresholds defined by pixel-
244 size quintiles, comparing mean inequalities for each threshold to those derived from the smallest
245 20% of pixels using 95% confidence intervals from bootstrapped distributions sampled with
246 replacement 10⁴ times (Table S4). We do not observe statistically significant differences in mean
247 daily TROPOMI inequalities outside of the 95% confidence intervals compared to the smallest
248 pixels. The lack of pixel area dependence suggests most city-wide NO₂ inequalities, and those that
249 are observed by TROPOMI, are driven by spatially clustered NO_x sources. TROPOMI pixels are
250 larger than the length scales of individual dispersion gradients; however, when NO_x sources are
251 clustered into source regions, their gradients also spatially aggregate. TROPOMI resolves NO₂
252 gradients on the scale of these source regions, if not individual sources, with the latter causing the
253 information loss compared to GCAS.

254 Observed NO₂ inequalities based on TVCDs are sensitive to the number of census tracts with NO₂
255 measurements across the domain (UA or MSA).⁴¹ When observation coverage is low, inequalities
256 tend to be based on TVCDs in census tracts less representative of city-wide demographics. In this
257 case, census tracts where high numbers of residents are in population groups in the majority with
258 respect to city area (not necessarily population count) are overrepresented in the calculation. The
259 net effect is that population-weighted inequalities are based on census tracts that have higher
260 populations of non-Hispanic whites than in the domain on average. In New York City–Newark,
261 Dressel et al.⁴¹ found low observation coverage biased NO₂ inequalities low by 6–7 percentage
262 points and, as a result, identified minimum coverage threshold requirements for daily mean NO₂

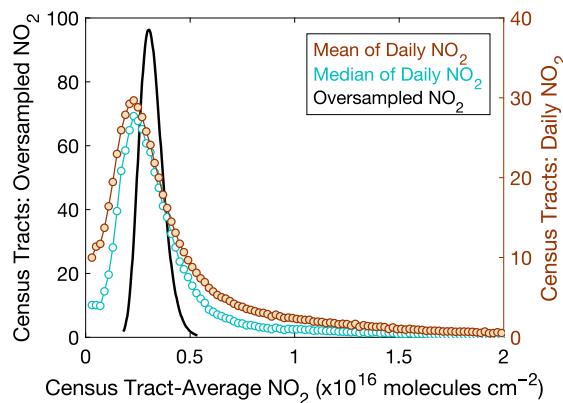
263 inequalities. We test sensitivity of mean daily TROPOMI NO₂ inequalities in Houston, first
264 applying a minimum coverage requirement of 20% of census tract with observations, then binning
265 daily TVCDs by >20%, >40%, >60%, and >80% census tracts with observations. When bootstrap
266 95% confidence intervals (calculated with replacement 10⁴ times) for a lower coverage bin do not
267 overlap with the 95% confidence interval for the >80% coverage bin, we identify a significant
268 difference between inequalities. We select thresholds separately for each metric as the lowest
269 coverage bin without a significant difference. Coverage thresholds range 20–40% for relative and
270 absolute inequalities for each metric (Table S5) and are applied throughout. Mean daily TROPOMI
271 NO₂ inequalities in Houston exhibit less observational coverage sensitivity than in New York
272 City–Newark.⁴¹

273 We compare mean daily NO₂ inequalities to results based on NO₂ TVCDs on the same subset of
274 days oversampled to 0.01° × 0.01° (~1 km × 1 km) using a physics-based algorithm⁸⁹ prior to
275 census tract averaging (Table 1). Oversampling averages measurements over time with large and
276 overlapping pixels to a finer grid, allowing sub-pixel-scale spatial features to be recovered.⁸⁹ The
277 oversampling approach used here treats pixel-level observations as sensitivity distributions using
278 a generalized two-dimensional super Gaussian spatial response function, appropriate for imaging
279 grating spectrometers like TROPOMI. Relative mean daily and oversampled NO₂ inequalities are
280 equal to within associated uncertainties; however, absolute NO₂ inequalities in mean daily TVCDs,
281 which are already low relative to fine-scale airborne remote sensing (Figure 2), are as much as
282 ~30% higher than oversampled TVCDs. We see multiple possible explanations for this:
283 oversampling is not enhancing spatial gradients relevant to describing census tract-scale NO₂
284 inequality, which is instead determined by the spatial resolving power set by pixel size; there is

285 limited NO₂ variability on scales of 1–4 km as relevant to NO₂ inequalities; and/or there is
286 compensating information in the daily inequalities lost through time averaging.

287 First, we compare NO₂ inequalities based on oversampled TVCDs over a range of grid sizes,
288 finding no significant differences in relative or absolute inequalities when we oversample to 0.01°
289 × 0.01°, 0.02° × 0.02°, 0.04° × 0.04° (the approximate TROPOMI nadir resolution), and 0.06° ×
290 0.06°. In an analysis of 52 major U.S. UAs, Demetillo et al.¹⁶ also reported small differences in
291 relative and absolute census tract-scale NO₂ inequalities using TROPOMI TVCDs oversampled to
292 0.01° × 0.01° and 0.04° × 0.04°, with the exceptions of the narrow coastal Californian cities of
293 Oakland, San Diego, and San Francisco, where NO₂ inequalities based on TVCDs oversampled to
294 0.04° × 0.04° were biased low by 8–22% compared to TVCDs oversampled to 0.01° × 0.01°,
295 suggesting oversampling enhances spatial gradients from coarser pixels when that variability
296 exists.¹⁶ Second, we take advantage of the natural variability in TROPOMI pixel orientations,
297 separately comparing NO₂ inequalities based on oversampled TVCDs to mean NO₂ TVCDs
298 collected within individual S-5P orbits, thus eliminating the oversampling pixel overlap
299 requirement. On average, for the 15 S-5P satellite orbits that fully cover the Houston UA, relative
300 NO₂ inequalities from oversampled and mean NO₂ TVCDs are similar; however, absolute NO₂
301 inequalities of mean TVCDs are ~30% higher than oversampled TVCDs for Black and African
302 Americans and Hispanics and Latinos (Table 1; Table S6), indicating the information loss is not
303 simply because of time averaging, but smoothing during oversampling. In Figure 3, we compare
304 mean and median distributions of tract-scale daily and oversampled (0.01° × 0.01°) TROPOMI
305 TVCDs, fit assuming distributions are lognormal as is characteristic for NO₂. Mean daily
306 measurements span a wider range of NO₂ conditions and retain more observations in the high tail
307 of the distribution than oversampled TVCDs, with high NO₂ values driving inequalities. Sun et

308 al.⁸⁹ report that oversampling, including with the physics-based algorithm used here, is more
 309 accurate when the grid is fine relative to a gradient with a smooth spatial response, for example, a
 310 city edge, while pixel means are more accurate for coarse grids and sharper spatial responses. Our
 311 results suggest absolute census tract-scale NO₂ inequalities are more accurately represented using
 312 means, with TROPOMI pixels and typical oversampling grids being large relative to scale of
 313 dispersion. Research using oversampled NO₂ TVCDs to identify NO_x point sources and infer NO_x
 314 emissions and NO₂ lifetimes have improved absolute estimates by rotating spatially variable NO₂
 315 plumes to a common wind direction,⁹⁰⁻⁹³ an aspatial solution not applicable to describing census
 316 tract-scale NO₂ inequalities, although potentially useful for informing related decision-making.



317
 318 **Figure 3.** Lognormal distributions of census tract-average TROPOMI NO₂ TVCDs in the Houston
 319 UA (May 2018–November 2022). Left axis: TVCDs oversampled to $0.01^\circ \times 0.01^\circ$ (black line).
 320 Right axis: mean (brown filled circles) and median (cyan open circles) of distributions of daily
 321 observations.

322 **Table 1.** Mean daily TROPOMI NO₂ inequalities at the MSA and UA level (May 2018–November
 323 2022) on days meeting observation coverage thresholds, inequalities based on TROPOMI NO₂
 324 TVCDs oversampled to $0.01^\circ \times 0.01^\circ$, $0.02^\circ \times 0.02^\circ$, $0.04^\circ \times 0.04^\circ$, and $0.06^\circ \times 0.06^\circ$, and average
 325 inequalities of the 15 TROPOMI orbit patterns that cover the Houston UA separately from means
 326 and oversampled TVCDs ($0.01^\circ \times 0.01^\circ$). Uncertainties are expressed as standard mean errors.

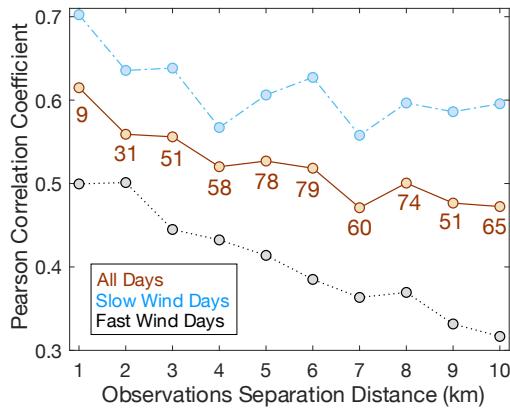
| Mean Daily TROPOMI | | | Oversampled TROPOMI | | | | Separately by TROPOMI Orbit | |
|---------------------------|----|-----|---------------------|---------------|---------------|---------------|-----------------------------|--|
| MSA | UA | MSA | UA | | | UA | | |
| | | | 0.01° × 0.01° | 0.02° × 0.02° | 0.04° × 0.04° | 0.06° × 0.06° | Mean | Oversampled ($0.01^\circ \times 0.01^\circ$) |
| Relative Inequalities (%) | | | | | | | | |

| | | | | | | | | | |
|---|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| Black and African Americans | 17 ± 1 | 8 ± 1 | 18 ± 1 | 9 ± 1 | 9 ± 1 | 8 ± 1 | 8 ± 1 | 9 ± 1 | 9 ± 1 |
| Hispanics and Latinos | 23 ± 1 | 16 ± 1 | 25 ± 1 | 17 ± 1 | 17 ± 1 | 17 ± 1 | 16 ± 1 | 18 ± 1 | 16 ± 1 |
| Asians | 9 ± 1 | -1 ± 1 | 11 ± 1 | 0 ± 1 | 1 ± 1 | 1 ± 1 | 2 ± 1 | 4 ± 1 | 2 ± 1 |
| Absolute Inequalities ($\times 10^{14}$ molecules cm $^{-2}$) | | | | | | | | | |
| Black and African Americans | 6.4 ± 0.5 | 3.6 ± 0.3 | 5.0 ± 0.3 | 2.7 ± 0.3 | 2.7 ± 0.3 | 2.6 ± 0.3 | 2.6 ± 0.4 | 3.7 ± 0.5 | 2.8 ± 0.4 |
| Hispanics and Latinos | 8.8 ± 0.5 | 6.8 ± 0.4 | 7.2 ± 0.4 | 5.4 ± 0.3 | 5.4 ± 0.3 | 5.3 ± 0.3 | 5.2 ± 0.4 | 7.3 ± 0.5 | 5.3 ± 0.4 |
| Asians | 3.7 ± 0.4 | 0.3 ± 0.4 | 2.9 ± 0.3 | 0.1 ± 0.3 | 0.1 ± 0.3 | 0.4 ± 0.4 | 0.5 ± 0.4 | 0.2 ± 0.5 | 0.4 ± 0.4 |

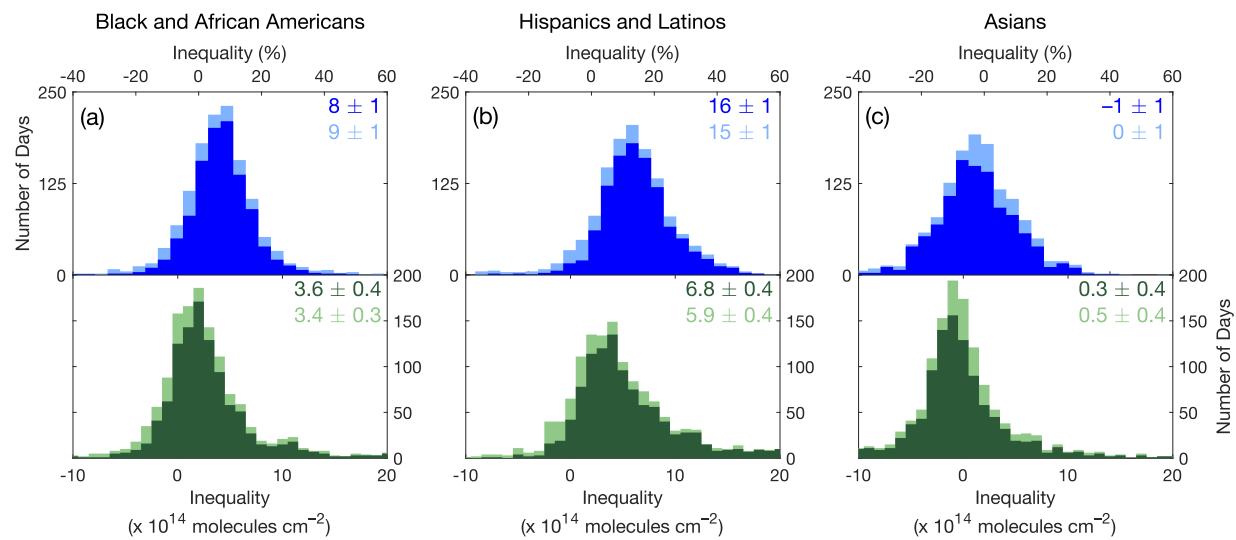
327

328 To describe spatiotemporal variability in column-surface relationships, we compare daily tract-
 329 average TROPOMI TVCDs and daytime (12–3 pm LT) NO₂* surface mixing ratios across the
 330 MSA as a function of their separation distance using Pearson correlation coefficients (r) over May
 331 2018–November 2022 (Figure 4).^{3, 16, 41, 69} We require NO₂* mixing ratio data at four or more
 332 monitors in each 1-km distance bin per day and exclude near-roadway monitors, which are subject
 333 to hyperlocal effects. Surface NO₂* and directly overhead TVCDs (defined as tract center points
 334 within 1 km of an NO₂* monitor) are strongly correlated, with median r values of 0.62. Correlation
 335 coefficients decrease as the distance between observations increases, falling to 0.54 on average
 336 when tract-average TVCDs are 2–6 km from the nearest monitor and 0.48 at 7–10 km. This r -
 337 distance dependence indicates spatial variability in daily TROPOMI TVCDs follows NO₂*
 338 patterns at the surface, with r decreases at 1–2 km consistent with length scales of NO₂ dispersion
 339 gradients. If we consider uncertainties as standard mean errors based on the number of days with
 340 observations included in the daily average, uncertainties in r are typically ±0.01 and mean
 341 differences in r with distance are significant. However, column-surface relationships are variable
 342 daily, with standard deviations (1σ) of ~0.3 in each distance bin. Daily correlation coefficients are
 343 lower than for oversampled TROPOMI TVCDs as reported in Demetillo et al.,³ especially at 1 km,
 344 meaning time averaging masks temporal variability in column-surface agreement. We also sort
 345 daily observations in the highest (>3.9 m s $^{-1}$) and lowest (<2 m s $^{-1}$) UA-wide mean daytime (12–3
 346 pm LT) surface wind quartiles as a function of distance, as wind is a physical control over the

347 inter-tract NO₂ distribution. Daily column-surface correlations covary with wind speeds physically
 348 realistically, with stronger *r* values for slower winds and smaller *r* values with faster winds at all
 349 observation separation distances.



350
 351 **Figure 4.** Median daily Pearson correlation coefficients between tract-averaged NO₂ TVCDs and
 352 surface NO₂* mixing ratios as a function of observation separation distance (km) on all days over
 353 May 2018–November 2022 (brown solid line) and on days in low (light blue dashed line) and high
 354 (black dotted line) quartile winds. We indicate the mean number of census tracts in the daily
 355 correlation at that distance each day, with similar statistics on low and high wind days.



356
 357 **Figure 5.** Daily UA-level TROPOMI NO₂ inequalities (May 2018–November 2022). Relative (%)
 358 and absolute (molecules cm^{-2}) inequalities on all days (light blue and light green, respectively) and
 359 on days meeting metric-specific coverage thresholds (bright blue and dark green, respectively) for
 360 Black and African Americans (a), Hispanics and Latinos (b), and Asians (c). Bootstrap mean
 361 inequalities, sampled with replacement 10^4 times, are reported with uncertainties as 95%
 362 confidence intervals.

363 **Daily NO₂ Inequalities.** We calculate daily TROPOMI census tract-average NO₂ inequalities over
364 May 2018–September 2022 across the Houston UA and MSA (Table 1; Figure 5). Mean daily UA-
365 level population-weighted NO₂ TVCDs are $8 \pm 1\%$ and $16 \pm 1\%$ higher for Black and African
366 Americans and Hispanics and Latinos compared to non-Hispanic/Latino whites, respectively.
367 Neighborhoods near the Houston Ship Channel (Figure 1) with large populations of Black and
368 African Americans and Hispanics and Latinos, e.g., Pasadena, Fifth Ward, Harrisburg/Manchester,
369 and Galena Park, often have the highest NO₂ concentrations. Mean population-weighted NO₂
370 TVCDs for each group including non-Hispanic/Latino whites are shown in Table S7. Inequalities
371 for Black and African Americans and Hispanics and Latinos increase to $17 \pm 1\%$ and $23 \pm 1\%$,
372 respectively, at the MSA level. Mean daily population-weighted NO₂ TVCDs for Asians equal
373 those for non-Hispanic/Latino whites within the UA but are $9 \pm 1\%$ higher across the MSA, mainly
374 due to the inclusion of the large Asian population around Sugar Land in southwest Houston (Figure
375 1d). We observe larger inequalities at the MSA level, reflecting urban-suburban differences,
376 compared to the UA, representing intraurban NO₂ differences.^{3, 94} UA and MSA-level relative (r
377 = 0.83–0.92) and absolute ($r = 0.88$ –0.95) inequalities are strongly correlated (Figure S3). Errors
378 for mean inequalities are 95% confidence intervals, which we derive from bootstrapped
379 distributions sampled with replacement 10^4 times. Absolute census tract-scale NO₂ inequalities are
380 often lower than the precision of individual TROPOMI NO₂ TVCDs, which have a median daily
381 pixel-level precision of 9.9×10^{14} molecules cm⁻² (approximately 30% of mean NO₂ TVCDs) over
382 May 2018–November 2022 in the Houston UA. However, this imprecision improves through
383 spatial and temporal averaging,⁹⁵ done here through population weighting over all census tracts in
384 the UA or MSA and by reporting daily inequality results as means over many days. Sampling and
385 nonsampling (e.g., measurement, coverage, nonresponse, and processing errors) errors in the ACS

386 influence the accuracy and precision of tract-scale NO₂ inequalities as well and, when random,
387 also improve through averaging to higher geographic levels.

388 We report NO₂ inequalities during 27 TRACER-AQ flights using GCAS separately in the late
389 morning, midday, and afternoon (Table 2). Relative inequalities are not statistically significantly
390 different with time of daytime, although there may be a tendency toward lower relative inequalities
391 at midday. Absolute NO₂ inequalities are significantly higher in the morning than midday and
392 afternoon, and there are multiple factors that could influence these differences. While wind speeds
393 are similar on average during all flights, the atmosphere is typically more stable in morning than
394 at midday, affecting the NO₂ distribution in the nearfield of NO_x sources,¹⁹ with convective mixing
395 common in the afternoon in Houston. The surface mixed layer height is typically shallower in the
396 morning than afternoon; however, this will have a larger effect on surface concentrations than
397 TVCDs. We also expect higher rush hour NO_x emissions and longer NO₂ chemical lifetimes⁹⁶ in
398 the morning and late afternoon compared to midday. Diurnal variability in absolute inequalities
399 has implications for interpreting observations from TROPOMI, which collects measurements at
400 12–3 pm LT over Houston, and the recently-launched TEMPO (Tropospheric Emissions:
401 Monitoring of Pollution) instrument, which scans North America hourly during daylight hours
402 from onboard a geostationary satellite.⁹⁷ Our analysis in the New York City–Newark UA found
403 fewer statistically significant morning-afternoon differences in absolute NO₂ inequalities,⁴¹
404 suggesting there is more to learn from TEMPO concerning temporal variability in the NO₂
405 distribution. Because GCAS subsampled the MSA, we also report mean daily TROPOMI NO₂
406 inequalities (May 2018–November 2022) along a representative TRACER-AQ flight for
407 comparison (Table 2).

408 **Table 2.** Relative and absolute mean GCAS NO₂ inequalities in the Houston MSA during
 409 TRACER-AQ in the morning (9–11:30 am LT), at midday (11:30 am–2 pm LT), and afternoon
 410 (2:30–5 pm LT). Relative and absolute mean daily TROPOMI NO₂ inequalities (May 2018–
 411 November 2022) along a representative TRACER-AQ flight raster (afternoon, 25 September
 412 2021). GCAS inequalities along spatially coincident TRACER-AQ and DISCOVER-AQ tracts
 413 during TRACER-AQ (2021) and DISCOVER-AQ (2013). Airborne and TROPOMI uncertainties
 414 are 95% confidence intervals of bootstrap mean inequalities, sampled with replacement 10⁴ times.

| | GCAS TRACER-AQ morning | GCAS TRACER-AQ midday | GCAS TRACER-AQ afternoon | TROPOMI along TRACER-AQ raster | 2021 GCAS (TRACER- AQ) | 2013 GCAS (DISCOVER- AQ) |
|---|------------------------------|-----------------------------|--------------------------------|--------------------------------------|------------------------------|--------------------------------|
| Relative Inequalities (%) | | | | | | |
| Black and African Americans | 17 ± 7 | 12 ± 6 | 13 ± 4 | 13 ± 1 | 9 ± 8 | 10 ± 6 |
| Hispanics and Latinos | 27 ± 4 | 20 ± 6 | 25 ± 2 | 22 ± 1 | 24 ± 6 | 20 ± 5 |
| Asians | 12 ± 10 | 13 ± 10 | 11 ± 4 | 3 ± 2 | 9 ± 6 | 11 ± 4 |
| Absolute Inequalities ($\times 10^{14}$ molecules cm ⁻²) | | | | | | |
| Black and African Americans | 14.4 ± 5.8 | 6.8 ± 3.9 | 8.2 ± 2.6 | 6.0 ± 0.7 | 4.9 ± 4.6 | 10.9 ± 6.3 |
| Hispanics and Latinos | 22.7 ± 4.4 | 11.9 ± 3.4 | 16.0 ± 3.6 | 10.6 ± 1.0 | 16.4 ± 3.7 | 19.3 ± 5.9 |
| Asians | 17.0 ± 12.9 | 9.2 ± 6.9 | 7.6 ± 3.8 | 2.5 ± 1.0 | 9.6 ± 6.2 | 9.4 ± 3.7 |

415
 416 GCAS NO₂ measurements in Houston collected during TRACER-AQ and DISCOVER-AQ offer
 417 observational insight into trends from 2013 to 2021 (Table 2). We compare weekday population-
 418 weighted, tract-average NO₂ columns in spatially coincident census tracts along representative
 419 TRACER-AQ and DISCOVER-AQ flight patterns (*SI Appendix 1*; Figure S4; Tables S8–S11).
 420 We calculate inequalities using the 2020 ACS for both DISCOVER-AQ and TRACER-AQ to
 421 allow comparisons across the same tracts and isolate effects of changes in NO₂ concentrations
 422 from demographics. We find relative NO₂ inequalities are statistically indistinguishable, with
 423 overlapping 95% confidence intervals for NO₂ inequalities in 2013 and 2021 and by the Wilcoxon
 424 rank sum test, a non-parametric two-sample t-test. While absolute inequalities were always lower
 425 during TRACER-AQ than DISCOVER-AQ, they were variable day to day, in addition to the
 426 relatively small number of aircraft observations, such that we lack the precision on their means
 427 (not the observations themselves) to interpret the differences. UA-wide mean NO₂* mixing ratios
 428 were slightly higher and more variable during DISCOVER-AQ (6.7 ± 6.2 ppb) than TRACER-
 429 AQ flights (6.0 ± 4.3 ppb); winds were slower during TRACER-AQ (2.1 ± 0.8 m s⁻¹) than

430 DISCOVER-AQ ($3.1 \pm 1.2 \text{ m s}^{-1}$). Slower mean winds during TRACER-AQ may have worsened
431 inequalities, while lower NO_2^* corresponds to lower absolute inequalities (discussed below).
432 Previous work has shown downward NO_x emissions trends have not reduced relative NO_2
433 inequalities in U.S. cities using NO_2 empirical models;^{12, 88} however, this has not yet been
434 demonstrated with observations directly to our knowledge.

435 Relationships between daily UA-level census tract-scale TROPOMI NO_2 inequalities, surface
436 winds, and overall NO_2 pollution (Table 3; Figures S5–S7) underscore the need for locally targeted
437 controls over sector-based approaches to reducing NO_2 disparities. Absolute NO_2 inequalities are
438 moderately negatively associated with wind speeds for most groups, as faster winds distribute NO_2
439 away from NO_x sources, showing NO_2 inequalities arise from the distribution of NO_x sources, as
440 well as that daily NO_2 inequalities vary meaningfully with relevant atmospheric conditions.
441 Absolute NO_2 inequalities moderately correlate with UA-mean surface NO_2^* and NO_2 TVCDs in
442 the winter and during O_3 season for most metrics. At the same time, relative inequalities are more-
443 weakly associated with overall NO_2 . Differences in these correlations for absolute and relative
444 NO_2 inequalities manifest from NO_x sources being systematically located in Black and African
445 American and Hispanic and Latino, as NO_2 concentrations in the nearfield of emitters are more
446 temporally variable than the physical locations of NO_x sources. As a consequence, emissions
447 reductions that maintain unequal source distributions, such as sector-based approaches, lower
448 overall NO_2 pollution and absolute differences between groups but have little effect on relative
449 inequalities, which require location-specific policy interventions.⁹⁸

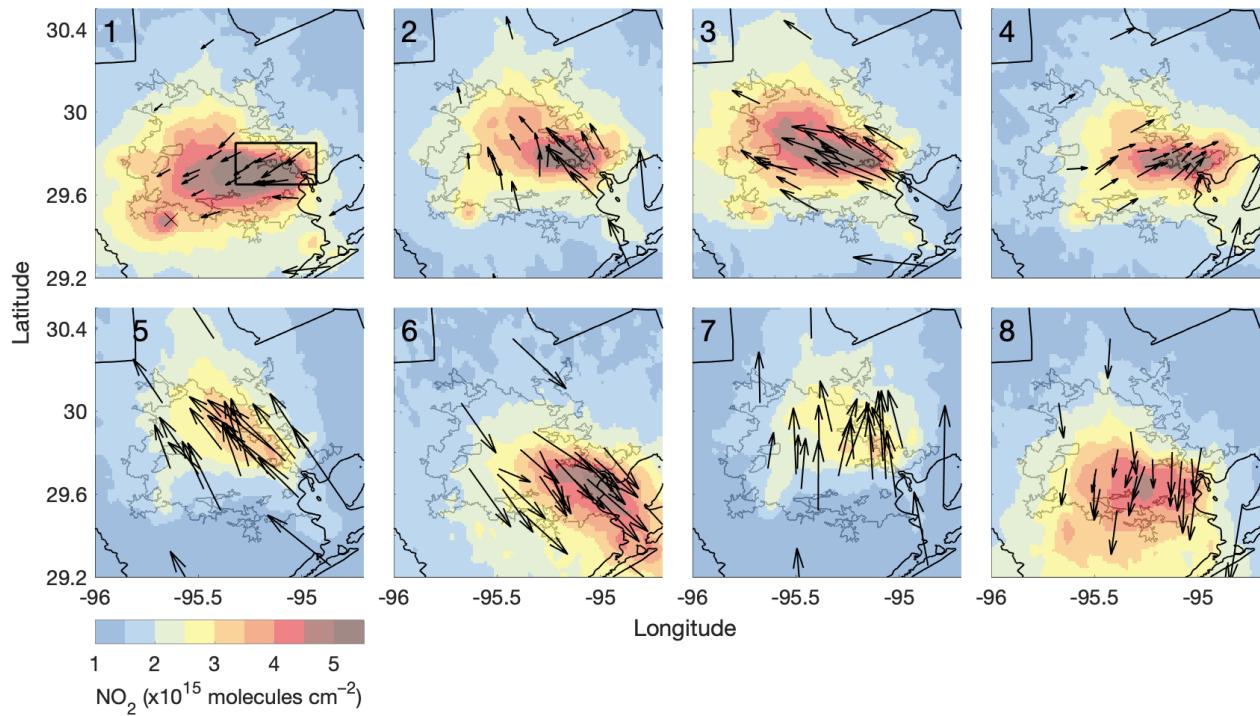
450 **Table 3.** Spearman rank correlation coefficients (2018–2022) with $p < 0.050$ in winter and O_3
451 season: daily absolute TROPOMI inequalities and daytime (12–3 pm LT) surface wind speed,
452 NO_2^* mixing ratios, and daily UA-level TROPOMI NO_2 TVCDs and daily relative TROPOMI
453 inequalities and daytime NO_2^* mixing ratios and UA-level TROPOMI NO_2 TVCDs.

| | Absolute Inequality Correlations | | | Relative Inequality Correlations | |
|-----------------------------|----------------------------------|---------------------------|-----------------------|--|-----------------------|
| | Wind Speed | Surface NO ₂ * | NO ₂ TVCDs | Surface NO ₂ * | NO ₂ TVCDs |
| | | | | Winter (December–February) | |
| Black and African Americans | −0.40 | 0.44 | 0.55 | 0.25 | 0.26 |
| Hispanics and Latinos | −0.62 | 0.67 | 0.67 | 0.31 | 0.17 |
| Asians | | | 0.21 | | 0.21 |
| | | | | O ₃ Season (March–November) | |
| Black and African Americans | −0.34 | 0.48 | 0.65 | 0.17 | 0.20 |
| Hispanics and Latinos | −0.51 | 0.61 | 0.77 | 0.24 | 0.26 |
| Asians | −0.17 | | 0.15 | | 0.07 |

454

455 **NO₂ Inequalities and O₃ Air Quality.** We use daily observations of NO₂ inequalities to
 456 investigate relationships between neighborhood-level NO₂ distributions and O₃ air quality. First,
 457 applying an established approach to understanding the influence of meteorology on O₃ variability
 458 in Houston, we disaggregate observations by winds using principal component and cluster
 459 analysis,^{48, 53, 99–103} presenting cluster characteristics that include census tract-scale NO₂
 460 inequalities. We generate one two-dimensional principal component for mean daytime (12–3 pm
 461 LT) u and v resultant winds during O₃ season, which captures 88% of the observed variability in
 462 u and v components. We then apply *k*-means clustering with 1,000 iterations to generate eight
 463 wind clusters, with the first centroid selected at random, from the iteration with the lowest total
 464 sum of distances (Figure 6; Table 4). We selected the optimal number of clusters, allowed to range
 465 1–10, using the Calinski-Harabasz criterion, maximizing the ratio of the between-cluster variance
 466 to the within-cluster variance with respect to the number of clusters.¹⁰⁴ We confirmed the identified
 467 number of clusters using the elbow method with 10³ iterations, with the optimal number of clusters
 468 based on the variance explained.¹⁰⁵ Eight clusters balances clarity and complexity relevant to
 469 relationships between NO₂ inequalities and MDA8 O₃. Missing daytime winds are filled using
 470 measurements from the closest proximity monitor with observations. We renamed the clusters 1–
 471 8 from most to least frequent MDA8 O₃ NAAQS exceedances. The analysis reproduces results in
 472 the literature, with high O₃ days associated with easterly and east-southeasterly winds.^{48, 53, 100, 103}

473 Figure 6 highlights the variability in NO₂ spatial distributions lost through averaging (Figure 1c),
474 with results based on long-term or annual averages representing conditions that infrequently occur.



475
476 **Figure 6.** Distinct mean daytime (12–3 pm LT) wind clusters during O₃ season (March–
477 November) over May 2018–November 2022 in the Houston MSA. Corresponding TROPOMI NO₂
478 TVCDs oversampled to $0.01^\circ \times 0.01^\circ$. Wind vector length is proportional to wind speed, with
479 mean wind speeds given in Table 4. The W.A. Parrish Generating Station is indicated with an \times
480 and the Houston Ship Channel with a thick black box in cluster 1. The thin inner gray and outer
481 black lines are the UA and MSA boundaries, respectively.

482 MDA8 O₃ NAAQS exceedances are most frequent in cluster 1, when winds are on average slow
483 and easterly—corresponding to the largest absolute daily TROPOMI race-ethnicity inequalities
484 (Table 4). Cluster 1 is the primary wind condition in which we observe statistically significant
485 UA-level inequalities for Asians, with NO₂ from the Ship Channel transported toward Sugar Land
486 in southwest Houston and stagnant NO_x emissions around the nearby coal-fired W.A. Parrish
487 Generating Station. This explains why NO₂ inequalities for Asians are not strongly correlated with
488 wind speed or overall NO₂ pollution level (Table 3). MDA8 O₃ NAAQS exceedances are also

489 common in clusters 2–4, when winds are slow ($\sim 1.6 \text{ m s}^{-1}$) and east-southeasterly, southerly, and
490 westerly, with elevated UA-level absolute daily TROPOMI NO_2 inequalities for all groups except
491 Asians. Clusters 5–8 include the fewest number of O_3 NAAQS exceedances, occurring on <5% of
492 days. These clusters are characterized by faster winds, lower UA-mean NO_2^* , and lower absolute
493 tract-scale daily TROPOMI NO_2 inequalities. Wind conditions have less influence on relative NO_2
494 inequalities, as winds do not affect the locations NO_x sources. Observed correspondence between
495 MDA8 O_3 and absolute census tract-scale NO_2 inequalities indicates similar atmospheric
496 conditions exacerbate both phenomena and/or high O_3 and NO_2 inequalities are linked chemically.

497 **Table 4.** Mean daytime wind cluster characteristics: number of days in each cluster; MSA-mean wind speed ($\pm 1\sigma$) and direction; MSA-
 498 level MDA8 O₃ NAAQS exceedances, both number and frequency; UA-mean NO₂* ($\pm 1\sigma$); and mean daily TROPOMI relative and
 499 absolute inequalities based on bootstrapped distributions sampled with replacement 10⁴ times with uncertainties as 95% confidence
 500 intervals.

| Cluster | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 |
|--|-----------|------------|-----------------------|------------|---------------|---------------|------------|---------------|
| Number of days | 186 | 279 | 193 | 112 | 170 | 48 | 162 | 136 |
| Wind speed (m s ⁻¹) | 0.7 | 1.2 | 2.2 | 1.3 | 3.7 | 4.2 | 3.5 | 2.5 |
| Wind direction | easterly | southerly | east southeasterly | westerly | southeasterly | northwesterly | southerly | northwesterly |
| Temperature (°C) | 28 | 32 | 27 | 30 | 28 | 23 | 30 | 23 |
| O ₃ NAAQS exceedances | 49 | 38 | 22 | 12 | 9 | 2 | 5 | 4 |
| O ₃ exceedance frequency (%) | 26 | 14 | 11 | 11 | 5 | 4 | 3 | 3 |
| NO ₂ * (ppb) | 7.4 | 6.6 | 7.2 | 6.1 | 5.1 | 5.5 | 3.9 | 6.7 |
| UA | | | | | | | | |
| Mean Daily Relative Inequalities (%) | | | | | | | | |
| Black and African Americans | 11 ± 1 | 13 ± 1 | 10 ± 2 | 9 ± 3 | 9 ± 2 | -3 ± 3 | 10 ± 1 | 6 ± 2 |
| Hispanics and Latinos | 19 ± 2 | 22 ± 2 | 17 ± 2 | 18 ± 6 | 14 ± 2 | 14 ± 3 | 13 ± 2 | 9 ± 2 |
| Asians | 9 ± 2 | 0 ± 1 | -1 ± 2 | -6 ± 2 | -3 ± 2 | -15 ± 4 | -0 ± 1 | 4 ± 2 |
| Mean Daily Absolute Inequalities ($\times 10^{14}$ molecules cm ⁻²) | | | | | | | | |
| Black and African Americans | 5.9 ± 1.1 | 5.0 ± 0.8 | 3.7 ± 0.6 | 3.3 ± 0.9 | 2.3 ± 0.6 | -0.7 ± 0.5 | 2.1 ± 0.3 | 2.1 ± 0.5 |
| Hispanics and Latinos | 9.5 ± 1.4 | 9.1 ± 1.4 | 6.9 ± 0.8 | 8.1 ± 1.5 | 3.7 ± 0.6 | 4.1 ± 0.8 | 2.9 ± 0.4 | 2.9 ± 0.5 |
| Asians | 5.3 ± 1.5 | -0.1 ± 0.5 | -0.4 ± 1.0 | -0.9 ± 1.4 | -0.7 ± 0.4 | -3.1 ± 0.5 | -0.0 ± 0.3 | 1.8 ± 0.7 |

501

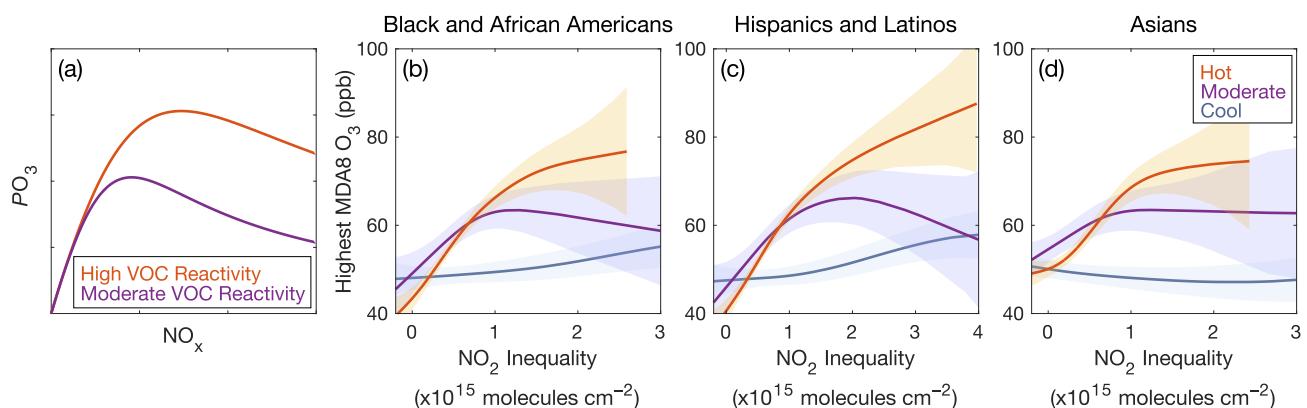
502 PO_3 varies nonlinearly with NO_2 concentrations (Figure 7a); therefore, NO_2 inequalities and city-
503 wide O_3 air quality are potentially coupled chemically. Briefly, PO_3 increases with increasing NO_x
504 when NO is the limiting reagent in O_3 -forming radical cycling (PO_3 chemistry is NO_x limited).
505 PO_3 decreases with increasing NO_x when NO_2 predominately combines with OH to produce nitric
506 acid, reducing O_3 -forming reactions between OH and VOCs (PO_3 is NO_x suppressed). This
507 nonlinear chemistry has important regulatory consequences, as NO_x decreases improve O_3 air
508 quality when chemistry is NO_x limited, while the same reductions worsen NO_x -suppressed O_3 .
509 When PO_3 dominates the O_3 mass balance, MDA8 O_3 varies as the integral of PO_3 across the
510 intraurban NO_2 heterogeneity, and, in Houston, NO_x -limited and suppressed conditions are both
511 present.⁵⁶ Because PO_3 depends nonlinearly on NO_2 , we describe O_3 -season relationships between
512 NO_2 inequalities and the highest daily MSA-level MDA8 O_3 using a generalized additive model
513 (GAM), a regression approach previously applied to nonlinear systems, including O_3 .¹⁰⁶⁻¹¹⁰ PO_3
514 also depends nonlinearly on VOC reactivity to OH, defined as the sum of the product of VOC
515 concentrations and their bimolecular reaction rate with OH.¹¹¹ Temperature is a proxy for VOC-
516 OH reactivity where a major portion of VOC emissions are temperature dependent, verifiable
517 through the observed O_3 - NO_2 dependence under different temperatures.¹¹² To consider VOC-OH
518 reactivity, we apply the GAM separately under low (<25°C), moderate (25–28°C), and high
519 (>28°C) daytime mean temperatures conditions. Results informing GAM selection and evaluation
520 are available in the Supporting Information (*SI Appendix* 2; Tables S12–13; Figures S8–S16).

521 GAMs of MDA8 O_3 versus NO_2 inequalities reproduce the nonlinear dependence of PO_3 on NO_2
522 concentrations (Figure 7). The highest MDA8 O_3 occur hot days, i.e., under higher VOC-OH
523 reactivity conditions, and when absolute NO_2 inequalities are large. We observe lower MDA8 O_3
524 when temperatures are moderate (lower VOC-OH reactivity) and NO_2 inequalities are large, with

525 similar MDA8 O₃ to hot days when NO₂ is more evenly distributed (PO_3 is NO_x limited). At low
526 temperatures, relationships between MDA8 O₃ and NO₂ inequalities suggest a more limited role
527 for PO_3 on MDA8 O₃. A key observation is that the transition between NO_x-limited and NO_x-
528 suppressed PO_3 chemistry that is near peak MDA8 O₃ occurs at higher absolute NO₂ inequalities
529 under higher temperature conditions, consistent with hotter temperatures corresponding to higher
530 VOC-OH reactivities, which in turn require more NO₂ to drive nitric acid production.¹¹² While at
531 very high NO concentrations O₃ can be titrated to NO₂, O₃ titration does not have the same
532 functional form as PO_3 with VOC-OH reactivity versus NO₂.

533 The GAMs demonstrate that NO₂ inequalities affect PO_3 chemistry and not merely that MDA8 O₃
534 and NO₂ inequalities covary under certain atmospheric conditions. We note, it is not the
535 inequalities per se, but the unequal NO₂ distributions resulting from NO_x sources being
536 disproportionately located in a subset of neighborhoods that drives PO_3 . That said, NO_x emission
537 sources overburden communities of color because of environmental racism in historical and
538 contemporary decision-making. Past research has already shown that PO_3 chemistry is spatially
539 heterogenous within Houston,^{46, 49, 56, 113, 114} and, because PO_3 chemistry is nonlinear, it follows
540 logically that the same NO_x emission reductions applied evenly across a city would be less
541 effective than a series of localized controls responsive to specific PO_3 mechanisms (NO_x limited
542 versus NO_x suppressed) as they vary in space. Wang et al.⁵⁸ used the adjoint of the Community
543 Multiscale Air Quality model focused on California to determine that PO_3 is disproportionately
544 sensitive to spatially localized controls. Our work implies that NO_x emissions controls that
545 eliminate neighborhood-level NO₂ inequalities will have O₃ air quality co-benefits, with regulatory
546 decision-making consolidating NO_x sources in a subset of Houston neighborhoods hindering O₃
547 NAAQS compliance. While MDA8 O₃ is largely NO_x limited with respect to NO₂ inequalities on

548 high temperature days, MDA8 O₃ is more NO_x suppressed as a function of NO₂ inequalities when
 549 temperatures are moderate, meaning even steeper NO_x reductions that also have the effect of
 550 decreasing NO₂ inequalities are required to lower O₃ under these conditions. Based on observed
 551 differences in correlations between absolute and relative NO₂ inequalities with overall NO₂ (Table
 552 3), decreases in NO₂ inequalities, and hence MDA8 O₃, require locally targeted NO_x reductions in
 553 neighborhoods where residents are primarily Black, Latinx, and Asian.



554
 555 **Figure 7.** Analytical model demonstrating relationships between PO_3 , NO_x , and VOC-OH
 556 reactivity (a). GAMs of daily MSA-level absolute TROPOMI NO_2 inequalities ($molecules\ cm^{-2}$)
 557 versus highest daily MDA8 O_3 (ppb) during O₃-season (March–November 2018–2022) on days
 558 meeting coverage thresholds under moderate (purple) and high (orange) daily maximum
 559 temperatures for Black and African Americans (b), Hispanics and Latinos (c), and Asians (d).
 560 Envelopes are 95% confidence intervals.

561 **Implications.** In Houston, daily TROPOMI NO_2 TVCDs capture a major portion of census tract-
 562 scale NO_2 inequalities compared to spatiotemporally coincident GCAS measurements that resolve
 563 length scales of dispersion. Mean daily TROPOMI NO_2 inequalities are insensitive to TROPOMI
 564 pixel size after the initial information loss with respect to GCAS. In Houston, and other U.S. cities,
 565 communities of color are statistically overburdened by air pollution sources,^{98, 115, 116} including
 566 NO_x sources.¹⁶ This is a consequence of historical (e.g., redlining) and contemporary (e.g.,
 567 permitting) decision-making that clusters emission sources in a subset of city neighborhoods,

568 creating source regions such as the Houston Ship Channel, in combination with historical and
569 contemporary policies and practices causing and reinforcing housing segregation,¹ including white
570 violence, housing discrimination, and separating communities with freeways.¹¹⁷⁻¹²¹ When NO_x
571 sources are in close proximity, their individual pollutant decay gradients also spatially aggregate;
572 as a result, a major portion of inequalities persist over spatial scales greater than length scales of
573 dispersion, the physical process motivating the application of very-high spatial resolution models
574 and measurements. Fine-scale observations are therefore not always required as evidence of air
575 pollution inequalities or to inform related policy making and accountability. While daily TVCDs
576 are coarse (20–89 km²), they retain a wider range of NO₂ values, especially in the high tail of the
577 NO₂ distribution, which drive inequalities. Daily mean NO₂ TVCDs result in higher, and therefore
578 more accurate, absolute NO₂ inequalities than oversampled TVCDs (0.01° × 0.01°), as TROPOMI
579 pixels and oversampling grids are large relative to the scale of dispersion. This has relevance to
580 future work based on TEMPO observations, which are not anticipated to meet the pixel overlap
581 requirements for oversampling.

582 We find that neighborhood-level NO₂ inequalities and city-wide O₃ are coupled air quality issues
583 in Houston. GAMs relating NO₂ inequalities and MDA8 O₃ under different temperature conditions
584 reproduce established nonlinear relationships between PO₃, NO₂, and VOC-OH reactivity. This
585 has policy consequences, producing empirical evidence that MDA8 O₃ is sensitive to the spatial
586 distribution of NO_x emissions reductions. O₃ control is typically approached through sector-based
587 NO_x and VOC emissions reductions without also considering distributive inequalities in O₃
588 precursors.¹²² However, we find that targeted NO_x emissions reductions where NO_x sources are
589 clustered—in communities of color—would lower both NO₂ inequalities and city-wide MDA8 O₃
590 in Houston, especially on hot days when MDA8 O₃ is highest. This means that permitting and

591 other policies concentrating sources in a subset of Houston neighborhoods affect O₃ NAAQS
592 attainment and calls for a reconceptualization of decision-making to include facility/emissions
593 location.

594 While there is growing evidence that locally-targeted regulatory interventions are required to
595 reduce and eliminate air pollution disparities,^{41, 98} there are barriers to their adoption, as
596 community-focused air quality plans and recommendations potentially cannot be pursued through
597 policy making at any level.¹²³ Houston and Pasadena (which is in the Houston UA) are among the
598 few major U.S. municipalities without formal zoning, an established tool for localities to influence
599 their own land use, including air pollution source distribution, through the institution of bans,
600 programs, and environmental review processes.¹²⁴ Additionally, Houston's efforts to address air
601 quality concerns through the local ordinance process have been invalidated by the Texas Supreme
602 Court,^{125, 126} further limiting the city from regulating emissions from facilities permitted by the
603 Texas Commission on Environmental Quality (TCEQ). TCEQ does not have an office or staff
604 focused on environmental justice, chooses not to use that term (any relevant activities are instead
605 described as Title VI compliance), and continues to issue permits without considering cumulative
606 impacts, including facility clustering. However, TCEQ does have a commitment to O₃
607 compliance,¹²⁷ making this a politically available pathway for addressing inequality in absence of
608 other approaches. Here, we demonstrate that MDA8 O₃ varies as a function of these neighborhood-
609 level NO₂ inequalities, with locally-targeted NO_x emissions controls required to address NO₂
610 disparities and having substantial O₃ air quality co-benefits. This conclusion has policy relevance
611 as the state has the authority, resources, and initiative to meet the O₃ NAAQS and is also evidence
612 that TCEQ must contend with practices and policies of environmental racism to improve O₃ air
613 quality.

614 **Supporting Information.** Surface wind roses corresponding the Figure 1, surface monitor
615 locations, detailed TRACER-AQ inequality results, population weighting equation, TROPOMI
616 inequalities as a function of observation coverage and pixel area, comparison of oversampled and
617 time-averaged inequalities by S-5P orbit, mean daily TROPOMI population-weighted NO₂,
618 correlations between daily UA and MSA-level inequalities, details for the comparison between
619 DISCOVER-AQ and TRACER-AQ inequalities, scatterplots of NO₂ inequalities versus surface
620 winds and NO₂*, and technical details on generalized additive model (GAM) construction,
621 including comparisons of GAM methods.

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631 Census database through the IPUMS National Historical Geographic Information System
632 (nhgis.org)¹²⁸ and TIGER/Line shapefiles of Texas census tract polygons and UA and MSA
633 boundaries from the Data.gov library (census.gov/cgi-bin/geo/shapefiles/index.php). NO₂*,
634 MDA8 O₃, and wind speed, wind direction, and temperature datasets were downloaded via the
635 U.S. EPA Air Quality System (aqs.epa.gov/aqsweb/documents/data_api.html).

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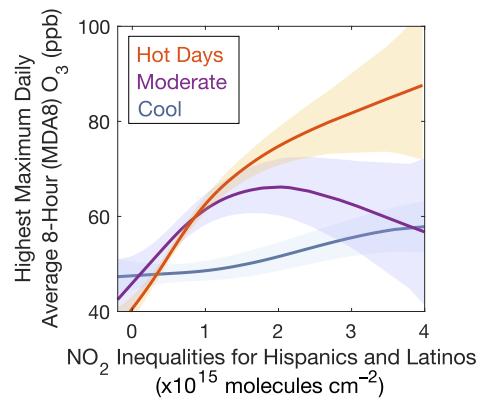
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